

Cascade-exciton model analysis
of proton spallation from 10 MeV to 5 GeV

S. G. Mashnik* and A. J. Sierk

T-2, Theoretical Division, Los Alamos National Laboratory, Los Alamos, NM 87545

O. Bersillon

CEA, Centre d'Etude de Bruyères-le-Châtel, 91680, Bruyères-le-Châtel, France

and

T. Gabriel

Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831

Abstract

We have used an extended version of the Cascade-Exciton Model (CEM) to analyze more than 600 excitation functions for proton induced reactions on 19 targets ranging from ^{12}C to ^{197}Au , for incident energies ranging from 10 MeV to 5 GeV. We have compared the calculations to available data, to calculations using approximately two dozen other models, and to predictions of several phenomenological systematics. We present here our conclusions concerning the relative roles of different reaction mechanisms in the production of specific final nuclides. We comment on the strengths and weaknesses of the CEM and suggest possible further improvements to the CEM and to other models.

*On leave of absence from Bogoliubov Laboratory of Theoretical Physics,
Joint Institute for Nuclear Research, Dubna, Russia

Precise nuclear data on excitation functions for reactions induced by nucleons in the energy range up to several GeV are of great importance both for fundamental nuclear physics and for many applications. Such data are necessary to understand the mechanisms of nuclear reactions, to study the change of properties of nuclei with increasing excitation energy, and to study the effects of nuclear matter on the properties of hadrons and their interactions. Excitation functions are more sensitive to the detailed mechanisms of nuclear reactions than are double differential cross sections of emitted particles or their integrals over energy and/or angles. Therefore, excitation functions are a convenient tool to test models of nuclear reactions.

Second, and perhaps more important today, expanded nuclear data bases in this intermediate energy range are required for several important applications. Recently, one of the most challenging problems requiring reliable nuclear data files is Accelerator-Driven Transmutation Technology (ADTT) for elimination of nuclear waste [1]. The problems of Accelerator Transmutation of Waste (ATW) are closely connected with Accelerator-Based Conversion (ABC) [2] aimed to complete the destruction of weapons plutonium, and with Accelerator-Driven Energy Production (ADEP) [3] which proposes to derive fission energy from thorium with concurrent destruction of the long-lived waste and without the production of weapons-usable material, though substantial differences among these systems do exist [2]. Precise nuclear data are needed for solving problems of radiation damage to microelectronic devices [4] and not only of radiation protection of cosmonauts and aviators or workers at nuclear installations, but also to estimate the radiological impact of radionuclides such as ^{39}Ar arising from the operation of fusion reactors or high-energy accelerators and the population dose from such radionuclides retained in the atmosphere so as to avoid possible problems of radiation health effects for the whole population (see, e.g. [5]). Another important new application which requires large nuclear data libraries at energies up to several hundreds of MeV is the radiation transport simulation of cancer radiotherapy used for selecting the optimal dose in clinical treatment planning systems [6]. Many excitation functions are needed for the optimization of commercial production of radioisotopes widely used in different branches of nuclear medicine [7], mining and industry [8]. Also, residual product nuclide yields in thin targets irradiated by medium- and high-energy projectiles are extensively used in cosmochemistry and cosmophysics, e.g. to interpret the production of cosmogenic nuclides in meteorites by primary galactic particles [9], etc.

Because of the impracticality of measuring all cross sections important to the processes of pragmatic interest, it is important to try to develop reliable models to predict cross sections which have not been or cannot be measured. In order to carefully evaluate the strengths and weaknesses of one such model, we have undertaken a careful comparison of an extended Cascade-Exciton Model [10] as realized in the CEM95 code with both experimental data on excitation functions for proton-induced reactions and with many other model calculations. We have studied the dependence of our results on the physics incorporated in the code, on the values of input parameters, on the incorporation of the isotopic composition of actual experimental targets,

and on the proper modeling of independent and cumulative yields.

We have performed detailed analyses of more than 600 excitation functions for interactions of protons with energies from 10 MeV to 5 GeV with nuclei of ^{12}C , ^{14}N , ^{16}O , ^{27}Al , ^{31}P , ^{40}Ca , ^{54}Fe , ^{56}Fe , ^{57}Fe , ^{58}Fe , ^{nat}Fe , ^{59}Co , ^{90}Zr , ^{91}Zr , ^{92}Zr , ^{94}Zr , ^{96}Zr , ^{nat}Zr , and ^{197}Au . We have compared our results with all reliable experimental data available to us and with predictions of other models realized in several codes: ALICE LIVERMORE 87 [11], HETC/KFA-2 [12], ALICE91 [13], LAHET [14], ALICE-F [15], NUCLEUS [16], MCEXCITON [17], ALICE82 [18], DISCA2 [19], CASCADE [20], HETC [21], INUCL [22], ALICE75 [23], ALICE LIVERMORE 82 [24], ALICE 87 MOD [25], PEQAQ2 [26], ALICE92 [27], CEM92M [28], with the Milan version of the exciton model of nuclear reactions with preformed α -clusters in nuclei [29], and with calculations using phenomenological systematics from Refs. [30]–[33]. A comparison of many of our results with predictions of several other codes may be found in a recent NEA/OECD document [34]. A comparison of the yields of residual product nuclei in ^{209}Bi thin targets irradiated by 130 MeV and 1.5 GeV protons simulated by CEM95 with the recent measurements by Titarenko et al. [35] and with results obtained with the codes HETC [21], GNASH [36], LAHET [14], INUCL [22], CASCADE [20], and ALICE96 [37] may be found above in this issue in the previous paper [35].

A detailed report of the study [38], containing 179 pages, 103 figures, and 243 references to 308 original books, journal articles, preprints, theses, and conference contributions is available on the World Wide Web as a compressed PostScript file, or in hard copy from either of the first two authors. Here we will present only our main conclusions from the study.

Our analyses have shown that several different mechanisms participate in the production of most final nuclides. Their relative roles change significantly with the changing atomic mass of the targets, with increasing incident energy, and are different for different final nuclides. The main nuclide production mechanism in the spallation region is the successive emission of several nucleons, while emission of complex particles is important (and may be even the only mechanism for production of a given isotope in a limited range of incident energy) only at low incident energies, near the corresponding thresholds, while with increasing energy its relative role decreases quickly.

For medium and especially for heavy targets, the contribution from radioactive precursors to the measured yields of many nuclides is very important. The cumulative yields of some nuclides are up to two orders of magnitude higher than the independent ones. Therefore, for heavy targets, especially careful calculations of cumulative yields and their comparisons with the measured data are needed.

Our analyses have shown that nuclear structure effects are very important in production of some nuclides and manifest themselves strongly even at an incident energy of 5 GeV. Therefore, reliable and well fitted models of shell and pairing corrections, level density parameters, and especially of nuclear masses and consequent binding energies and Q -values have to be used in calculations.

The extended version of the cascade-exciton model realized in the code CEM95

describes satisfactorily with a fixed set of input parameters the shapes and absolute values of the majority of measured excitation functions for production of nuclides in the spallation region and for the emission of secondary nucleons and complex particles. We feel that the yields of both nuclides in the spallation region and secondary particles of $A < 4$ predicted by CEM95 are at least as reliable, and in many cases more so, than those of the other models and phenomenological systematics mentioned above.

For target nuclei from ^{27}Al to ^{197}Au , CEM95 describes the majority of experimental excitation functions in the spallation region to within a factor of 2. For targets lighter than ^{27}Al , the agreement with experimental data is worse, and the CEM, like the majority of other models, has to be improved to be able to describe excitation functions from light targets. Because CEM95 does not contain a special mechanism for fragmentation, because it underestimates production of ^4He , and does not include a model of fission fragment production, it cannot reliably predict nuclide yields in the mass and energy regions where these processes are dominant. These mechanisms of nuclear reactions will need to be incorporated into the CEM.

In rare cases, in the same spallation region where it is usually reliable, CEM95 underestimates or overestimates some individual measured excitation functions, sometimes up to an order of magnitude. This is mainly a result of the poor nuclear mass and binding energy values used in CEM95.

We conclude that the extended version of the cascade-exciton model realized in the code CEM95 is suitable for a rough evaluation of excitation functions in the spallation region. But for a better description of the measured yields in this region and for an extension of the range of its applicability into the fission and fragmentation regions, it should be developed further. Among improvements of the CEM which are of highest priority we consider the following:

- incorporation of recent experimental nuclear mass tables, and new reliable theoretical mass formulas for unmeasured nuclides,
- development and incorporation of an appropriate model of high-energy fission,
- modeling the emission of gammas competing with the evaporation of particles at the compound stage,
- treating more accurately α -emission at the preequilibrium stage,
- incorporation of a model for fragmentation of medium and heavy nuclei, and the Fermi breakup model for highly excited light nuclei,
- modeling the evaporation of fragments with $A > 4$ from not too light excited nuclei (incorporation of such processes at the preequilibrium stage may also be important),
- modeling the coalescence of light fragments from fast emitted particles,

- improvement of the approximations for inverse cross sections, and
- use of new, more precise experimental data for the cross sections of elementary interactions at the cascade stage.

Such a development and improvement of the CEM is possible, and work in this direction is already in progress. We hope that a proper incorporation of the above improvements in the code will not destroy the present wholeness of CEM95 and its good predictive power for the spectra of secondary particles.

There are a number of other possible and desirable improvements of the CEM discussed in the complete report [38], which are justified from a physical point of view. Unfortunately, the inclusion of separate refinements in nuclear models used in INC calculations does not always lead to improved agreement with experimental data.

The problems discussed above are typical not only of the CEM, but also for all other similar models and codes, where they are also not solved yet. Excitation functions are a very “difficult” characteristic of nuclear reactions as they involve together the different and complicated physics processes of spallation, evaporation, fission, and fragmentation of nuclei. A lot of work is still necessary to be done by theorists and code developers before a reliable complex of codes able to satisfactorily predict arbitrary excitation functions in a wide range of incident energies/projectiles/targets/final nuclides will be available. At present, we are still very far from the completion of this difficult task.

In the meantime, to evaluate excitation functions needed for science and applications, it is necessary to use and analyze together the available experimental data, and for each region of incident energies/projectiles/targets/final nuclides, the predictions of phenomenological systematics and the results of calculations with the most reliable codes. Our present study has shown that for proton-induced reactions in the spallation region, not too low incident energies and not too light targets, CEM95 is such a reliable code.

Acknowledgements

It is a pleasure to acknowledge M. Blann, V. P. Eismont, L. M. Krizhansky, R. Michel, P. Nagel, H. S. Plendl and Yu. E. Titarenko for useful discussions on spallation physics which have stimulated us to carry out this research. One of the authors (S. G. M.) thanks CEA, Bruyères-le-Châtel and ORNL, Oak Ridge for kind hospitality and excellent conditions during his work in February–August 1996 at Bruyères-le-Châtel and in July–August 1995 at Oak Ridge, where most of this study was done. He is also grateful to M. B. Chadwick, R. E. MacFarlane, D. G. Madland, P. Möller, J. R. Nix, R. E. Prael, L. Waters, and P. G. Young of LANL for fruitful discussions and support.

This study was completed under the auspices of the U.S. Department of Energy by the Los Alamos National Laboratory under contract no. W-7405-ENG-36.

References

- [1] Second Int. Conf. on Accelerator-Driven Transmutation Technologies and Applications, Kalmar, Sweden, June 3–7, 1996; Proc. Int. Workshop on Nucl. Methods for Transmutation of Nuclear Wastes: Problems, Perspectives, Cooperative Research, Dubna, Russia, May 29–31, 1996, M. Kh. Khankhasaev, Zh. B. Kurmanov and H. S. Plendl, eds., World Scientific, Singapore (1997); Overview of Physics Aspect of Different Transmutation Concepts, NEA / NSC / DOC (94) 11, OECD Nuclear Energy Agency (1994); J. P. Schapira, Transmutation of Nuclear Wastes, A working report to Nupecc, I. P. N. Report IPNO DRE 94-04, Orsay (1994) and references therein.
- [2] C. D. Bowman, in: Proc. Int. Workshop on Nucl. Methods for Transmutation of Nuclear Wastes: Problems, Perspectives, Cooperative Research, Dubna, Russia, May 29–31, 1996, M. Kh. Khankhasaev, Zh. B. Kurmanov and H. S. Plendl, eds., World Scientific, Singapore, (1997) p. 257 and references therein.
- [3] C. Rubbia, in: Proc. Int. Conf. Nuclear Data for Science and Technology, May 9-13, 1994, Gatlinburg, TN, USA, J.K. Dickens, ed., American Nuclear Society, Inc., La Grange Park, IL (1994) p. 1065; C. Rubbia, J. A. Rubio, S. Buono, F. Carminati, N. Fiétier, J. Galvez, C. Gelès, Y. Kadi, R. Klapisch, P. Mandrillon, J. P. Revol, C. Roche, CERN Report CERN/AT/95-44(ET), (1995); K. D. Tolstov, JINR Rapid Communications, 5 [62] (1993) 5 and JINR Report 18-92-303, Dubna (1992); I. V. Chuvilo, G. V. Kiselev, B. R. Bergelson, B. P. Kochurov, in: Proc. Int. Conf. and Technol. Exposition on Future Nucl. Systems: Emerging Fuel Cycle and Waste Disposal Options, GLOBAL'93, Seattle, Washington, September 12–17, 1993, (1995) p. 924; V. P. Dmitrievskii, Fiz. Elem. Chastits At. Yadra 28 (1997) 815 [Phys. Part. Nucl. 28 (1997) 322].
- [4] V. S. Barashenkov, A. N. Sosnin, S. Yu. Shmakov, N. G. Goleminov, A. Polanski, Fiz. Element. Chastits At. Yadra 24 (1993) 246 [Phys. Part. Nucl. 24 (1993) 107]; C. J. Gelderloos, R. J. Peterson, M. E. Nelson and J. F. Ziegler, University of Colorado Report NPL-1139, Boulder, CO, USA (1997).
- [5] K. Kitao, in: Proc. of the 1991 Symposium on Nuclear Data, November 28–29, 1991, JAERI, Tokai, Japan, M. Baba and T. Nakagawa, eds., JAERI-M, 92-027, INDC(JPN)-157/L, JAERI (1992) p. 249.
- [6] R. M. White, M. B. Chadwick, W. P. Chandler, C. L. Hartmann Siantar, C. K. Westbrook, in: Proc. Int. Conf. Nuclear Data for Science and Technology, May 9-13, 1994, Gatlinburg, TN, USA, J.K. Dickens, ed., American Nuclear Society, Inc., La Grange Park, IL (1994) p. 1023.
- [7] A. Hashizume, in: Proc. Int. Conf. on Nucl. Data for Science and Technology, May 30–June 3, 1988, Mito, Japan, S. Igrasi, eds., JAERI (1988) p. 1067;

- S. N. Dmitriev and N. G. Zaitseva, *Fiz. Elem. Chastis At. Yadra*, 27 (1996) 977 [*Phys. Part. Nucl.* 27 (1996) 403]; G. F. Stein, B. R. S. Simpson, S. J. Mills and F. M. Nortier, *Appl. Radiat. Isot.* 43 (1992) 1323.
- [8] E. A. Edmonds, in: *Radioisotope Techniques for Problem-Solving in Industrial Process Plants*, J. S. Charlton, ed., Leonard Hill, Glasgow (1986) p. 247.
 - [9] R. Michel, I. Leya and L. Borges, *Nucl. Instr. Meth. B*, 113 (1996) 434 and references therein.
 - [10] K. K. Gudima, S. G. Mashnik and V. D. Toneev, *Nucl. Phys. A* 401 (1983) 329.
 - [11] M. Blann, Private Communication (1987); cited in B. Dittrich, U. Herpers, H. J. Hofmann, W. Wölfl, R. Bodermann, M. Lüpke, R. Michel, P. Dragovitsch and D. Filges, *Nucl. Instr. and Meth. B* 52 (1990) 588.
 - [12] P. Cloth, D. Filges, R. D. Neef, G. Sterzenbach, C. Reul, T. W. Armstrong, B. L. Colburn, B. Anders and H. Brückmann, KFA Report KFA-IRE-EAN/12/88, Jül-2203 (1988).
 - [13] M. Blann, private communication (1991).
 - [14] R. E. Prael and H. Lichtenstein, Los Alamos National Laboratory Report LA-UR-89-3014 (1989); R. E. Prael and M. Bozoian, Los Alamos National Laboratory Report LA-UR-88-3238 (1988); many documents concerning LAHET are available on the World Wide Web at: <http://www-xdiv.lanl.gov/XTM/>.
 - [15] T. Fukahori, JAERI-M 92-039 (1992) 114.
 - [16] T. Nishida, Y. Nakahara and T. Tsutsui, JAERI-M-86-116 (1986).
 - [17] N. Kishida and H. Kadotani, *Proc. Int. Conf. on Nucl. Data for Science and Technology*, Mito, Japan, S. Igrasi, Ed., JAERI (1988) p. 1209.
 - [18] M. Blann and T. A. Komoto, Lawrence Livermore National Laboratory Report UCID-19390 (1982).
 - [19] A. Yu. Konobeev, Yu. A. Korovin and V. N. Sosnin, *J. Nucl. Materials* 186 (1992) 117; *Kerntechnik* 57 (1992) 188.
 - [20] V. S. Barashenkov, Le Van Ngok, L. G. Levchuk, Zh. Zh. Musulmanbekov, A. N. Sosnin, V. D. Toneev and S. Yu. Shmakov, JINR Preprint P2-85-173, Dubna (1985).
 - [21] T. W. Armstrong and K. C. Chandler, *Nucl. Sci. Eng.* 49 (1972) 110.

- [22] G. A. Lobov, N. V. Stepanov, A. A. Sibirtzev and Yu. V. Trebukhovski, ITEP Preprint ITEP-91, Moscow (1983); A. A. Sibirtzev, N. V. Stepanov and Yu. V. Trebukhovski, ITEP Preprint ITEP-129, Moscow (1985); N. V. Stepanov, ITEP Preprint ITEP-81, Moscow (1987); N. V. Stepanov, ITEP Preprint ITEP-55, Moscow (1988).
- [23] M. Blann, University of Rochester Report CCO/3494-29 (1975).
- [24] M. Blann, cited in R. Michel, F. Peiffer and R. Stück, Nucl. Phys. A 441 (1985) 617.
- [25] M. Blann and V. P. Lunev, Lawrence Livermore National Laboratory Reports UCID-19614, UCID-20169; International Code Comparison for Intermediate Energy Nuclear Data, M. Blann, H. Gruppelaar, P. Nagel and J. Rodens, eds., NEA OECD, Paris (1994) p. 161.
- [26] E. Betak, Report INDC(CSR)-016/LJ, IAEA, Vienna (1989); Report FU SAV 89/5, Inst. Phys., Bratislava (1989).
- [27] M. Blann, Lawrence Livermore National Laboratory Report UCRL-JC-109050, (1991).
- [28] S. G. Mashnik, in: Proc. Specialists' Mtg. on Intermediate Energy Nuclear Data: Models and Codes , Issy-les-Moulineaux, France, May 30–June 1, 1994, OECD, Paris (1994) p. 107.
- [29] E. Gadioli, E. Gadioli Erba and J. J. Hogan, Phys. Rev. C 16 (1977) 1404.
- [30] G. Rudstam, Z. Naturforsch. A 21 (1966) 1027.
- [31] B. K. Gupta, S. Das and M. M. Biswas, Nucl. Phys. A 155 (1970) 49.
- [32] M. Foshina, J. B. Martins and O. A. P. Tavares, Radiochim. Acta 35 (1984) 121.
- [33] A. Yu. Konobeyev and Yu. A. Korovin, Nucl. Instr. and Meth. B 82 (1993) 103.
- [34] R. Michel and P. Nagel, International Code and Model Intercomparison for Intermediate Energy Activation Yields, NSC/DOC(97)-1, NEA/P&T No 14, OECD, Paris (1997); <http://www.nea.fr/html/science/pt/ieay>.
- [35] Yu. E. Titarenko, O. V. Shvedov, M. M. Igumnov, R. Michel, S. G. Mashnik, E. I. Karpikhin, V. D. Kazaritsky, V. F. Batyaev, A. B. Koldobsky, V. M. Zhivun, A. N. Sosnin, R. E. Prael, M. B. Chadwick, T. A. Gabriel, M. Blann, Nucl. Instr. and Meth. A, the previous paper in this issue.
- [36] P. G. Young, E. D. Arthur and M. B. Chadwick, Los Alamos National Laboratory Report LA-12343-MS (1992); M. B. Chadwick and P. G. Young, Phys. Rev. C 47 (1993) 2255.

- [37] M. Blann, Phys. Rev. C 54 (1996) 1341.
- [38] S. G. Mashnik, A. J. Sierk, O. Bersillon and T. Gabriel,
Los Alamos National Laboratory Report LA-UR-97-2905 (1997);
<http://t2.lanl.gov/publications/publications.html>.